Probing the Single-Electron Energy Spectrum of Electrodeposited Semiconductor Q-dots

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Combining a regular crystal structure with a discrete energy spectrum, semiconductor nanocrystals (Q-dots, 1 to 10 nm in size) form a class of materials in between macroscopic solids and individual atoms [1]. Since they make up possible components of future (opto)electronic nanodevices, their optical and electronical properties have been investigated intensively over the last decade [2].

In general, Q-dots are formed by colloidal synthesis [2]. As demonstrated by various authors, the electronic properties of colloidal Q-dots may be studied by attaching them to a conducting substrate and probing them with an STM tip (tunneling spectroscopy) [3]. However, to probe the single-electron energy spectrum of a Q-dot, this double-barrier tunnel junction should be designed carefully. Firstly, the energy levels of either the tip or the substrate must remain fixed relative to the energy levels of the Q-dot [3b]. More important, charging of the Q-dot must be avoided since electron-electron interactions obscure the single-electron energy spectrum [3a].

In this work, we have attempted to measure the single-electron energy spectrum of electrodeposited PbS Q-dots. Here, the direct contact between the Q-dot and the conducting substrate ensures that tunneling of electrons from the dot to the substrate is much faster than tunneling from the tip to the dot. In this way, electron-electron interactions are avoided.

An STM-picture of electrodeposited PbS dots on Au(111) is shown in Fig. 1. The dots are approximately 4 to 5 nm in size. The corresponding I-V curve measured at a single PbS Q-dot at 4.2 K is shown in Fig. 2. A zero current gap of about 2.5 eV is found. Since bulk PbS has a bandgap of 0.41 eV, electrons in PbS Q-dots are strongly confined. This is also apparent from the absorption spectrum of colloidal PbS dots of approximately the same size (6 nm) as shown in Fig, 3, as it has a first absorption peak at 2.2 eV. At positive bias, a current step at about 1 V is observed in the I-V curve (Fig. 2), indicating resonant tunneling through the first electronic orbital. A doublet is not observed, thus confirming that electron-electron interactions are effectively avoided. The current increase at 1.9 V suggests a second resonance at much higher energy. This is in correspondence with the widely separated peaks in the absorption spectrum.

In summary, we have demonstrated very strong size quantization in electrodeposited PbS quantum dots. Therefore, this system will be very useful for a detailed study of electron confinement effects as a function of the size and the shape of the nanocrystals.

[1] L.E. Brus, *J. Chem. Phys.* **1984**, 80, 4403
[2] A.N. Shipway, E. Katz, I. Willner, *ChemPhysChem* **2000**, 1, 18
[3] a) U. Banin, Y.W. Cao, D. Katz, O. Milo, *Nature* 1999, 400, 542; b) E.P.A.M. Bakkers, D. Vanmaekelbergh, *Phys. Rev. B* **2000**, 62, R7743

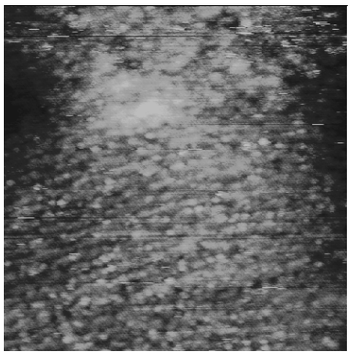

Fig. 1. STM-picture of PbS Q-dots (light) electrodeposited on a Au(111) surface (dark); scale: 120×120 nm. Dot size: 4 to 5 nm.

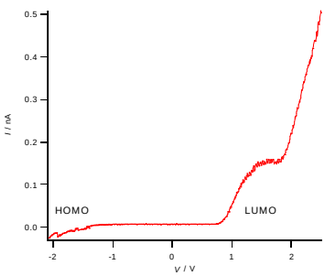

Fig. 2. I-V curve measured with a LT-STM (4.2 K) at a single PbS Q-dot electrodeposited on a Au(111).

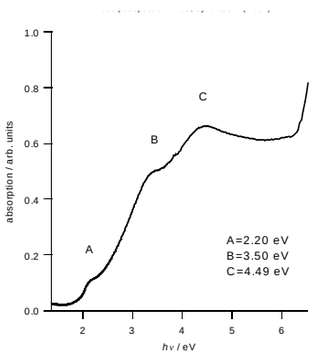

Fig. 3. Absorption spectrum of colloidal PbS Q-dots (diameter 6 nm).

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